Femtosecond laser detection of xenon and Stark decelerated polyatomic molecules

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Summary

The structure and properties of molecules make them invaluable test beds for exploring physics beyond the Standard Model. Precision measurements in molecules are used, for instance, to look for a possible time variation of fundamental constants such as the fine-structure constant and the proton-to-electron mass ratio, to test if fundamental laws obey time-inversion symmetry and to measure the energy difference between the two enantiomers of chiral molecules. Ultimately the resolution of any precision measurement is limited by the time a molecule spends in the measuring device. At VU LaserLab, we exploit the interaction of polar molecules with time-varying electric fields to produce beams of slow and cold molecules which can be used to test fundamental physics theories with unprecedented precision. For the success of this technique it is imperative that sensitive detection methods are developed that can also be applied to larger and heavier molecules.

In this thesis, a fs laser is employed for the first time to detect Stark decelerated and trapped molecules by non-resonant multiphoton ionization. The advantages of this detection scheme is twofold: (i) fs laser pulses have peak powers on the order of $10^{14}\,\mathrm{W/cm^2}$ at its focus. These high intensities are sufficient to ionize virtually any atom or molecule, thus potentially providing a very general and sensitive detection method for atoms and molecules. In particular, this method can be applied to polyatomic molecules, which in general lack suitable transitions for commonly used detection schemes such as resonance-enhanced multiphoton ionization (REMPI) or laser induced fluorescence (LIF), thus greatly enhancing the scope of molecules that can be Stark decelerated. (ii) as many (infrared) photons are required to ionize these molecules, the interaction volume is inherently restricted to the central part of the laser focus. This implies a small and well defined detection volume which allows for an accurate determination of the absolute densities of molecules. Moreover, this volume can be calibrated by simultaneously measuring the ion yield of a well-studied atom such as xenon.

Compared to resonant detection schemes, the obvious disadvantage of the fs-laser detection scheme is that it does not provide any state-specificity, which complicates the interpretation of the experiment and gives an increased background. However, in typical Stark-deceleration experiments, rotational cooling

in the supersonic expansion combined with state selection in the Stark decelerator ensure that only one or a few states are populated in the decelerated beam. Furthermore, in this thesis we demonstrate that background signals from thermal gas in our chamber can be effectively suppressed by using velocity map imaging (VMI) and mass selection, hence this non-resonant detection scheme using a fs laser can be quite useful for the detection of Stark-decelerated molecules.

This thesis consists of four chapters, which describe four different experiments. Chapter 2 adopts the fs ion imaging technique to characterize the density profile of pure and seeded molecular beams generated from the home-built cantilever piezovalve. The measured xenon beam density is two orders of magnitude lower than expected which is attributed to the severe skimmer clogging. Chapter 3 explores the feasibility of performing high resolution microwave spectroscopy on methanol in a Stark-deflected molecular beam. The motivation behind this work is that microwave transitions in methanol are demonstrated to be very sensitive to a possible variation of the proton-to-electron mass ratio. Unfortunately, no suitable resonant transitions are available for state-selective detection of methanol. We investigate if non-resonant detection using a fs laser in combination with an electric deflection field will be sufficiently sensitive and specific. In Chapter 4, non-resonant multiphoton ionization with a fs laser is used to detect Stark decelerated and trapped methyl fluoride molecules. Methyl fluoride is so far the heaviest and most complex molecule that has been decelerated to rest. It is demonstrated that the background resulting from thermal gas in the chamber can be effectively suppressed by the VMI technique. In addition, owing to the highly non-linear process, it turned out to be straight forward to accurately determine the density and the total number of trapped methyl fluoride molecules. Finally, in Chapter 5, the fs laser is used to detect Stark decelerated ammonia molecules and compare it to the conventional detection method that employs resonant multiphoton ionization by UV laser light from a ns dye-laser. The motivation for this work was the hope to increase the number of detected molecules, thereby increasing the precision of spectroscopic studies on cold ammonia. It is found that a fs laser shows no apparent advantage for the planned spectroscopy experiments on ammonia, partly because the experiment runs at a 10 Hz repetition rate while the femtosecond laser runs at 1 kHz. If all intensity of the fs laser could be used for the detection, the signal obtained with the fs detection scheme would be 100 times more than that of the ns detection scheme.